

## A POLARIZATION METHOD OF A MULTI-LAYERED PIEZOELECTRIC BODY

### BACKGROUND OF THE INVENTION

#### 1. Field of the Invention

The present invention relates to a polarization method of a multi-layered piezoelectric body used for a filter of a portable telephone, or other suitable electronic component, and more particularly, the present invention relates to a polarization method of a multi-layered piezoelectric body in which a plurality of piezoelectric layers and a plurality of internal electrodes are alternately laminated and adjacent piezoelectric layers are polarized in the thickness direction such that the polarization directions thereof are opposite to each other.

#### 2. Description of the Related Art

Conventionally, a length mode piezoelectric resonator has large design freedom, small spurious vibrations, and the difference  $\Delta f$  between a resonance frequency and an anti-resonance frequency is large. See, for example, Unexamined Japanese Patent Publication No. 10-4330 gazette.

Fig. 1 shows an example of this length mode piezoelectric resonator 10. The piezoelectric resonator 10 includes a base 11 in which a plurality of piezoelectric layers 12 and a plurality of internal electrodes 13 are laminated alternately. The piezoelectric layers on both sides of the internal electrodes 13 are polarized in opposite directions. Insulating films 14 and 15 are alternately provided to cover ends of the internal electrodes 13. Furthermore, external electrodes 16 and 17 are provided on opposing surfaces of the piezoelectric resonator 10. Therefore, the external electrodes 16 and 17 are alternately connected to every other one of the internal electrodes 13.

In the piezoelectric resonator 10, the polarization degree of the piezoelectric layer 12 greatly influences the properties thereof. Therefore, variations in the polarization degree within each element and variations in the polarization degree variation between elements must be minimized.

In this type of piezoelectric resonator, a block-like multi-layered piezoelectric body is provided. After polarization is performed, the piezoelectric body is cut into separate piezoelectric resonators. The polarization process of the multi-layered piezoelectric body is performed by the

method shown in Fig. 2. A multi-layered piezoelectric body 1 is defined by a block-like piezoelectric ceramic material member. Here, although four piezoelectric layers 1a to 1d are shown to simplify the explanation, many layers are laminated together to produce the piezoelectric resonator. Between the piezoelectric layers 1a to 1d, internal electrodes 2a to 2c are provided. The internal electrodes 2a to 2c are alternately led out to a side surface of the piezoelectric body 1, and are connected with side surface electrodes 3 and 4. Also, by applying the DC electric field between the side surface electrodes 3 and 4, as illustrated by arrow P, the piezoelectric layers 1b and 1c on both sides of the internal electrode 2b are polarized in opposite directions thereby obtaining a desired polarization degree.

However, in the method as shown in Fig. 2, because an electric field concentrates on the edge portion of the internal electrodes 2a to 2c, the polarization degree distribution is not uniform. Fig. 3 shows an example of the polarization degree distribution in one piezoelectric layer. An oblique line illustrates the polarization degree. As shown in the Fig. 3, if the electric field is applied in the thickness direction to the piezoelectric body 1, the polarization degree at the four corner sections of the piezoelectric body 1 is substantially increased (concave distribution), and a uniform polarization degree distribution is not obtained. As a result, when lamination elements used to form the block having piezoelectric layers with non-uniform polarization degree distributions are laminated, and the lamination is cut into a rectangular shape to define an element, it is impossible to use the peripheral sections of the piezoelectric body, and thus the yield of the piezoelectric body is greatly reduced.

For example, when performing the polarization of a multi-layered piezoelectric body for series resonators ( $f_r=450\text{kHz}$ ,  $f_d=55\text{kHz}$ ) used for a ladder-type filter by the method shown in Fig. 2, the variation in the polarization degree  $f_d$  in the block is at least  $10\text{kHz}$ . Therefore, only elements cut from near the center of the block can be used, and the polarization of the peripheral element of the block is defective and cannot be used.

Consequently, the inventions of the present application have suggested a method in which an electric field is applied to opposing external electrodes of a main surface of a multi-layered piezoelectric body, after performing the polarization (initial polarization) in one thickness direction of the multi-layered piezoelectric body, a side surface electrode which leads an internal electrode out is alternately provided. An electric field is applied between the side

electrodes and only the direction of polarization of the piezoelectric layer of one side of the internal electrode is reversed (polarization reversal), and a desired polarization degree is obtained. See, for example, Japanese Unexamined Patent Application No. 2000-52743. In this method, as shown in Fig. 4, even when there is a variation in polarization degree  $\Delta P1$  between a peripheral section and a center section in the initial polarization, when the electric field is applied in the opposite direction and the direction of polarization is reversed, polarization degree variations are reduced to  $\Delta P2$ . Thus, the non-uniformity of the polarization degree distribution after an initial polarization is corrected.

However, when the polarization of a saturated polarization degree  $P_{max}$  of the piezoelectric layer having a direction of polarization that is reversed is performed until it becomes almost equal to saturated polarization degree  $P_{max}$  at the time of an initial polarization, although the polarization degree variation is reduced, a polarization degree distribution of the piezoelectric layer in which polarization reversal is performed becomes concave in a similar manner as before the polarization reversal. Therefore, when a multi-layered piezoelectric body is constructed in which the piezoelectric layers are polarized in a reverse direction are laminated alternately by the above-mentioned method, the piezoelectric layer with a concave distribution in which the polarization reversal is performed and the piezoelectric layer with a concave distribution in which the polarization reversal is not performed are laminated alternately. Also, in a multi-layered piezoelectric body as a whole, a uniform polarization degree distribution is not reliably obtained.

#### SUMMARY OF THE INVENTION

To overcome the above-described problems, preferred embodiments of the present invention provide a polarization method for a multi-layered piezoelectric body that produces a polarization degree distribution of the entire multi-layered piezoelectric body that is uniform, thus greatly improving the yield.

According to a preferred embodiment of the present invention, a polarization method for a multi-layered piezoelectric body includes alternately laminating a plurality of piezoelectric layers and a plurality of internal electrodes and polarization of the adjacent piezoelectric layers is performed in the thickness direction thereof so as to polarize the adjacent piezoelectric layers in opposite directions, a first polarization process in which an electric field in one thickness direction is applied to the multi-layered piezoelectric body and

uniformly performs the polarization in the thickness direction, and a secondary polarization process in which the electric field is applied in the opposite thickness direction to the piezoelectric layers on both sides of the internal electrodes and only the direction of polarization of the piezoelectric layer of one side of the internal electrode is reversed. The above secondary polarization is performed in a range such that a remaining polarization degree  $Pr_2$  that exists after the secondary polarization in the piezoelectric layer having a direction of polarization that is reversed does not exceed a remaining polarization degree  $Pr_1$  that exists after the first polarization.

Further, according to a preferred embodiment of the present invention, in a polarization method of the multi-layered piezoelectric body in which a plurality of piezoelectric layers and a plurality of internal electrodes are laminated alternately and the polarization of the adjacent piezoelectric layers is performed in opposite thickness directions, a first polarization process is performed in which electric fields of opposite directions are applied to the piezoelectric layers on both sides of the internal electrode and the polarization is performed on the piezoelectric layers on both sides of the internal electrode in opposite directions, and a secondary polarization process is provided in which an electric field in the opposite direction of the electric field in the first polarization process is applied in opposite directions and the direction of polarization of the piezoelectric layers of both sides of the internal electrode is reversed, wherein the above secondary polarization is performed in a range such that the remaining polarization degree  $Pr_2$  that exists after the secondary polarization in the piezoelectric layer having a direction of polarization that is reversed does not exceed the remaining polarization degree of  $Pr_1$  that exists after the first polarization.

A first polarization is performed by applying an electric field in one thickness direction to a multi-layered piezoelectric body, to uniformly polarize the multi-layered piezoelectric body in the thickness direction. Next, a secondary polarization is performed by applying an electric field in an opposite direction relative to the piezoelectric layers on both sides of the internal electrode and only the direction of polarization of the piezoelectric layer of one side of the internal electrode is reversed.

Other features, elements, characteristics and advantages of the present invention will become more apparent from the following detailed description of the preferred embodiments with reference to the attached drawings.

## BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1 is a perspective diagram of an example of a piezoelectric resonator according to a preferred embodiment of the present invention.

Fig. 2 is a diagram showing a polarization method of the conventional multi-layered piezoelectric body.

Fig. 3 is a perspective diagram showing a polarization degree distribution of a block-like piezoelectric body in which a polarization was performed by the method of Fig. 2.

Fig. 4 is a diagram showing the polarization degree distribution at a time of the initial polarization and the reversed polarization.

Fig. 5 is a process drawing showing an example of a polarization method in accordance with a preferred embodiment of the present invention.

Figs. 6(a) to 6(d) are diagrams showing variations in the polarization degree distribution of two adjacent piezoelectric layers when performing the polarization method shown in Fig. 5.

Fig. 7 is a process drawing showing an example of a polarization method according to a preferred embodiment of the present invention.

Figs. 8(a) to 8(d) are diagrams showing variations in a polarization degree distribution of adjacent two piezoelectric layers when performing the polarization method shown in Fig. 7.

Fig. 9 is a diagram showing variations in the polarization degree of the reversed polarization layer when changing the applied voltage of a secondary polarization.

Fig. 10 is a diagram showing the polarization degree distribution in each point of A-F of Fig. 9.

Figs. 11(a) to 11(c) are process drawings showing an example of the polarization method in accordance with the first example of preferred embodiments of the present invention.

Figs. 12(a) to 12(d) are process drawings showing an example of the polarization method in accordance with the second example of preferred embodiments of the present invention.

Figs. 13(a) to 13(c) are process drawings showing an example of the polarization method in accordance with the third example of preferred embodiments of the present invention.

## DETAILED DESCRIPTION OF PREFERRED EMBODIMENTS

Fig. 5 shows an example of a polarization method according to a preferred embodiment of the present invention. First, front and back electrodes 5 and 6 are provided on front and back surfaces of the multi-layered piezoelectric body 1, the DC electric field in the thickness direction is applied to the multi-layered piezoelectric body 1, and the polarization (first polarization) is performed in one thickness direction. Then, the internal electrodes 2a to 2c are alternately led out to the outside surface of the multi-layered piezoelectric body 1 and connected to side surface electrodes 3 and 4. By applying the DC electric field between the side surface electrodes 3 and 4, electric fields of opposite directions are applied to the piezoelectric layers 1b and 1c on both sides of the internal electrode 2b and only the direction of polarization of the piezoelectric layer 1b of one side of the internal electrode 2b is reversed (secondary polarization). In addition, repolarization of only the piezoelectric layer 1c on the other side of the internal electrode 2b is performed. Thus, the direction of polarization is not reversed.

The polarization conditions at the time of the secondary polarization were changed, and the inventor discovered that a polarization degree distribution of the piezoelectric layer 1b having the reversed polarization axis varies with the degree (the remanent polarization degree) of progress of the secondary polarization.

Figs. 6(a) to 6(d) show the changes in the degree of distribution of polarization in the two piezoelectric layers 1b and 1c as the secondary polarization is performed. An arrow shows the direction of the polarization.

Fig. 6(a) shows the polarization degree distribution after the first polarization. The polarization degree distribution includes concave distributions. Figs. 6(b) to 6(d) show a variation of the polarization degree distribution as the secondary polarization is performed. In addition, Pr1 shows a remaining polarization degree caused by the first polarization. Pr2 shows a remaining polarization degree caused by the secondary polarization.

As clearly shown in Fig. 6, the polarization degree distribution of the piezoelectric layer 1b is a convex-shaped or flat distribution such as (b) and (c) when the direction of polarization is initially reversed. However, as shown in (d), the polarization degree distribution changes to a concave shape as the secondary polarization progresses. In addition, the polarization degree distribution of piezoelectric layer 1b in (b) and (c) is convex-shaped in the

example shown in Fig. 6. However, some materials have an approximately flat distribution. Thus, if the secondary polarization progresses too much, the polarization degree distribution of the piezoelectric layer 1b becomes a similar concave distribution as produced by the first polarization (however, the direction of a polarization is reversed). In addition, the polarization degree distribution of the piezoelectric layer 1c by remains concave.

Therefore, as shown in (d), when the reversed polarization progresses too much, the polarization degree distribution of the piezoelectric layer 1b in which the reversal polarization is performed and a polarization degree distribution of the piezoelectric layer 1c in which reversed polarization is not performed are both concave. Thus, a uniform polarization degree distribution in the overall piezoelectric body is not produced.

Consequently, the secondary polarization is terminated in a range such that the polarization degree distribution of the piezoelectric layer 1b having a direction of polarization that is reversed is either convex-shaped or flat. In other words, the secondary polarization is performed in a range wherein the remaining polarization degree  $Pr_2$  that exists after the secondary polarization does not exceed the remaining polarization degree  $Pr_1$  that exists after the first polarization, such that  $Pr_1 \geq Pr_2$ .

When the above equation is satisfied, since the polarization degree distribution of the piezoelectric layer 1b in which the direction of polarization is reversed is convex or flat and the polarization degree distribution of piezoelectric layer 1c in which the direction of polarization is not reversed is concave, the convex-shaped polarization degree distribution and the concave polarization degree distribution offset each other. Further, the degree of unevenness of the concave distribution is greatly reduced. Thus, the multi-layered piezoelectric body achieves a substantially uniform polarization degree distribution. As a result, the usable portion of the piezoelectric body greatly increases, and therefore the yield greatly improves.

In addition, where  $Pr_1 > Pr_2$ , the amplitude of the polarization degree of the two layers 1b and 1c is unbalanced. However, when the polarization degree distribution is substantially uniform, the resonance characteristic of a length vibration mode element is not adversely affected.

Where  $Pr_2 \doteq Pr_1$ , as shown in (c) of Fig. 6, the size of the polarization degree of the two layers 1b and 1c is approximately equal, and the distribution of the multilayer piezoelectric body is uniform, resulting in greatly improved

properties.

According to a second preferred embodiment of the present invention, a first polarization process includes a first process to apply an electric field in a first thickness direction to a multi-layered piezoelectric body, and a second process to apply the electric field in the thickness direction that is opposite to the first thickness direction to the multi-layered piezoelectric body. The direction of polarization of the multi-layered piezoelectric body produced by the first process is uniformly reversed by the second process.

Thus, the first polarization is performed only once. However, the polarization degree distribution is concave after the initial polarization of Fig. 4. Also, the difference  $\Delta P1$  between a center section and an edge portion is large. Consequently, multiple operations of the first polarization are performed. If a direction of polarization is reversed over the entire multi-layered piezoelectric body, the difference  $\Delta P2$  between a center section and an edge portion decreases when polarization reversal occurs of Fig. 4. Also, the non-uniformity of the degree distribution of polarization is corrected. Thus, the non-uniformity of the polarization degree distribution of the piezoelectric layer after the secondary polarization is corrected by correcting the non-uniformity of the degree distribution of polarization in the first polarization.

In addition, the second process may be performed multiple times.

According to a third preferred embodiment of the present invention, a first polarization that applies an electric field in opposite directions to the piezoelectric layers on both sides of the internal electrode to produce polarization of the piezoelectric layers on both sides of the internal electrode in opposite directions is performed. A secondary polarization in which electric fields in the opposite directions to the above-mentioned electric fields are applied to the piezoelectric layers on both sides of the internal electrode and the polarization axes of the piezoelectric layers on both sides of the internal electrode are reversed. In other words, the secondary polarization axial direction of the piezoelectric layers on both sides of the internal electrode is reversed relative to the first polarization axial direction.

Fig. 7 shows an example of the polarization method described above. First, the internal electrodes 2a to 2c are alternately led out to the outside surface of the multi-layered piezoelectric body 1. Side electrodes 3 and 4 are electrically connected to the internal electrodes 2a to 2c. In addition, the polarization is performed on the piezoelectric layers 1b and 1c on both sides of



the internal electrode 2b in opposite directions by applying a DC electric field between the side surface electrodes 3 and 4 (first polarization). This polarization process is the same as the polarization process in the prior art (Fig. 2). Next, a DC electric field in a reversed direction is applied between the side surface electrodes 3 and 4, and the polarization axes of the piezoelectric layers 1b and 1c on both sides of the internal electrode 2b are reversed simultaneously (secondary polarization).

Also in this case, the secondary polarization is performed in a range such that the remaining polarization degree  $Pr_2$  existing after the secondary polarization in the piezoelectric layers 1b and 1c having polarization axes that are reversed does not exceed remaining polarization degree  $Pr_1$  existing after the first polarization.

In this case, because the polarization axes of the piezoelectric layers 1b and 1c on both sides of the internal electrode 2b are reversed and a repolarization layer is not present, the polarization conditions of the two layers 1b and 1c are approximately equal. While attaining equalization of a polarization degree distribution, the amplitude of the polarization degree of the two layers 1b and 1c is also approximately equal.

Fig. 8 shows a change of the polarization degree distribution in the piezoelectric layers 1b and 1c when performing the secondary polarization by the method shown in Fig. 7. An arrow shows the direction of the polarization.

Fig. 8(a) shows the polarization degree distribution after the first polarization. The polarization degree distribution is concave. Figs. 8(b) to 8(d) show a variation of the polarization degree distribution when the secondary polarization is performed. After the direction of polarization is reversed by the secondary polarization, the degree of polarization of the two layers 1b and 1c is approximately equal, and the polarization degree distribution becomes flat or convex shaped. The degree of unevenness of the degree distribution of polarization of both the layers 1b and 1c is greatly reduced. If the secondary polarization is performed until the remaining polarization degree  $Pr_2$  is approximately equal to the remaining polarization degree  $Pr_1$  caused by the first polarization, as shown in (c), the two layers 1b and 1c have a flat or convex-shaped distribution, the polarization degree increases, and polarization degree distribution is optimized. If the secondary polarization progresses further, the distribution becomes concave as shown in (d).

Therefore, a multilayer piezoelectric body 1 having a uniform polarization

degree distribution is obtained by setting the secondary polarization conditions such that  $Pr1 \geq Pr2$ .

It is desirable to perform the first polarization process on a block-like multi-layered piezoelectric body, and to perform the secondary polarization process to the multi-layered piezoelectric body having a substantially rectangular shape.

That is, to enhance productivity, it is preferable to perform both the first polarization and the secondary polarization on a block-like multi-layered piezoelectric body. However, the non-uniformity of the polarization degree distribution at the time of the first polarization is three-dimensional, as shown in Fig. 3. Therefore, if the secondary polarization is performed on the block-like multi-layered piezoelectric body, the non-uniformity of the polarization degree distribution cannot be eliminated.

Consequently, if the secondary polarization is performed after cutting the block-like piezoelectric body into a substantially rectangular shape, the electric field strength and the duration can be set depending upon the polarization degree distribution of each substantially rectangular body. Therefore, variations in the degree of polarization between different substantially rectangular bodies and in the substantially rectangular body is greatly reduced.

In the multi-layered piezoelectric body in which the first polarization is performed, a length vibration is not excited because the polarization is uniformly performed in the thickness direction.

Consequently, a difference DF between the resonance frequency and an antiresonance frequency of an area expansion mode is calculated, and the degree of polarization is determined. On the other hand, in the multi-layered piezoelectric body in which the secondary polarization is performed, the length mode vibration is excited because the body includes layers in which the directions of polarization are different. The difference df between the resonance frequency and the antiresonance frequency of length mode is calculated, and then the polarization degree is determined. Because two types of polarization degrees having different vibration modes cannot be compared, DF of an area expansion vibration of the multi-layered piezoelectric body after the first polarization is converted into electro-mechanical-coupling-coefficient K, and set to a remaining polarization degree Pr1. The difference df of a length vibration of the multi-layered piezoelectric body after the secondary polarization is converted into electro-mechanical-coupling-coefficient K, and set to a

remaining polarization degree  $Pr_2$ . By comparing these remaining polarization degrees  $Pr_1$  and  $Pr_2$ , the amount of the secondary polarization is determined.

Moreover, in the third preferred embodiment, the multi-layered piezoelectric body in which the first polarization is performed includes a layer having a reversed polarization direction. Therefore, a length vibration is excited. Thus, both the remaining polarization degree  $Pr_1$  of a first polarization and the remaining polarization degree  $Pr_2$  of a secondary polarization can be calculated from the value which is attained by converting the difference  $df$  between the resonance frequency and antiresonance frequency of a length vibration into electro-mechanical-coupling-coefficient  $K$ .

Fig. 9 shows variations in the polarization degree  $Pr_2$  of a substantially rectangular layer having a direction of polarization that is reversed when the voltage applied in the second polarization is changed. The remaining polarization degree  $Pr_1$  is approximately equal to 50kHz in the first polarization for a desired PZT type piezoelectric ceramics. Fig. 10 shows the polarization degree distribution in each point A to F of Fig. 9. In addition, the change in the degree of polarization of the piezoelectric layer (a regular polarization layer) when changing an applied voltage is also illustrated in Fig. 9. The thickness of a piezoelectric layer is preferably about 0.56 mm.

Clearly from Figs. 9 and 10, in the layer in which the polarization is reversed, depolarization is performed in accordance with a voltage increase in a secondary polarization. A direction of polarization is reversed when the voltage is about 900v, and a polarization degree increases after that. If the voltage of a secondary polarization increases to about 1000v, after polarization reversal, the remanent polarization degree  $Pr_2$  of a polarization inversion layer will be set to be about 50kHz, and will become approximately equal to the remanent polarization degree of  $Pr_1$  of a first polarization. Until the voltage of the secondary polarization increases to near 900v to 1000v (referring to points D and E), the polarization degree distribution is approximately flat or slightly convex-shaped. When the voltage increases to more than about 1000v (referring to point F), the polarization degree distribution becomes concave.

Therefore, the voltage of the secondary polarization is set such that  $Pr_1 \geq Pr_2$ , in other words, the voltage is set to be in the range of about 900v to about 1000v.

In addition, the polarization degree is approximately 0 in a regular

polarization layer until the voltage exceeds about 500v. However, the polarization degree increases at voltages greater than about 500v. During that period, the polarization degree distribution within a rectangle is concave and it does not vary substantially.

In Figs. 9 and 10, although the piezoelectric layer in which the repolarization is performed is not illustrated, in the case of a repolarization layer, the electric field of the secondary polarization does not vary until the electric field in the secondary polarization exceeds the polarization degree in the first polarization. When the electric field in the secondary polarization exceeds the polarization degree of the first polarization, the polarization degree in the secondary polarization becomes greater than the polarization degree of the first polarization for the first time. During that period, the polarization degree distribution within a rectangle is still concave in the regular polarization layer and does not vary substantially.

Various aspects of preferred embodiments and comparative examples of the polarization method of the multi-layered piezoelectric body according to the present invention are explained below. In this example, a PZT type multi-layered piezoelectric body was used as a material of a length mode piezoelectric resonator ( $df=55\text{kHz}$ ).

#### First Example

Fig. 11 shows a manufacturing process of the length mode piezoelectric resonator according to a first example of preferred embodiments of the present invention.

First, an electro-conductive paste for internal electrodes that includes silver, palladium, an organic binder, or other suitable materials, is applied on one side of a green sheet that includes a piezoelectric ceramics, and these sheets are alternately laminated. The laminated structure is integrally baked at about 1200 degree C, and the multi-layered piezoelectric body 1 having a block shape with approximate dimensions of 20 mm x 30 mm x 3.9 mm is formed. Front and back electrodes 5 and 6 are provided on front and back surfaces of this block 1, and a DC electric field is applied between the front and back electrodes 5 and 6, and the first polarization is performed (referring to (a) of Fig. 11).

The conditions of the first polarization are as follows: electric field is about 1.5 kv/mm, polarization time is about 10 min, and retention-temperature is about

70 degrees C. After that, an aging process is performed at about 150 degrees Celsius for about 1 hour.

Next, side electrodes 3 and 4 leading out an internal electrode are alternately provided on a side surface of the block-like multi-layered piezoelectric body 1 after the first polarization. Then, this multi-layered piezoelectric body 1 was cut in a vertical direction to produce one substantially rectangular element using a dicer. To the rectangle 1A, a DC electric field was applied to the side surface electrodes 3 and 4, and the secondary polarization was performed (referring to (b) of Fig. 11). At this time, the polarization degree of each rectangle 1A was set to a desired value by controlling the polarization time. The conditions of the secondary polarization are set as follows: electric-field is 1.5 kv/mm, polarization temperature is 70 degrees C.

The polarization time is controlled to adjust the desired polarization degree (a polarization degree of the rectangle responded to the polarization degree of a length mode element,  $df=55\text{kHz}$ ). After that, the aging process was performed at about 250 degree Celsius for about 1 hour.

The range of the secondary polarization is determined to be as described in the following. The range, wherein the value in which the difference  $df$  between the resonance frequency and the antiresonance frequency of the length vibration mode of the rectangle 1A after the secondary polarization is converted into an electro-mechanical-coupling-coefficient  $K$  (remanent polarization degree  $Pr_2$ ), does not exceed the value of the difference  $DF$  between the resonance frequency and the antiresonance frequency of the area expansion oscillation mode of the block 1 after the first polarization is converted into an electro-mechanical-coupling-coefficient  $K$  (remanent polarization degree  $Pr_1$ ), that is,  $Pr_1 \geq Pr_2$ .

Every other electrode of the rectangle 1A exposed at the side surface was coated with an insulating material after the secondary polarization, and a silver electrode was provided thereon. This was cut by the dicer and a  $1.5\text{ mm} * 1.5\text{ mm} * 3.8\text{ mm}$  length mode piezoelectric resonator 1B was obtained (referring to (c) of Fig. 11).

Since the specific structure of this piezoelectric resonator 1B is the same as that of Fig. 1, the explanation thereof is omitted.

#### Comparative Example

A block-like multi-layered piezoelectric body is formed by a similar method

as the first example. Side surface electrodes for alternately leading out an internal electrode are provided on the side surface of the body. A DC electric field was applied to the side surface electrode of the multi-layered piezoelectric body, and the polarization was performed (referring to Fig. 2).

Polarization conditions are as follows: electric-field is 1.5 kv/mm, and retention-temperature is 70 degree C. A polarization time is controlled to achieve a desired polarization degree  $DF=2.0\pm 0.2\text{kHz}$ . Polarization degree  $DF$  was calculated from the difference between the resonance frequency and the antiresonance frequency of the area expansion oscillation mode of a block.

After that, the aging process was performed at about 250 degree C for about 1 hour, the piezoelectric block is cut into a desired size, and a length mode piezoelectric resonator was obtained.

The frequency characteristics of the impedance of the two elements which obtained by the above method are measured, and the value  $df=55\text{kHz}$  was measured as the difference between a resonance frequency and an antiresonance frequency.

Table 1 and Table 2 show the comparison in the polarization degree  $df$  and in the resonant frequency  $fr$  between the first example and Comparative Example in a property classification process, wherein  $\sigma_{n-1}$  is a standard deviation and  $r$  is the difference between the maximum value and the minimum value.

**Table 1**  
**First Example**

		Evaluation Lot 1	Evaluation Lot 2	Evaluation Lot 3	Average Value
df	Average	56.15	55.82	55.03	55.67
	$\sigma_{n-1}$	0.90	0.92	0.90	0.91
	max.	58.5	58.5	58.5	58.50
	min.	54	53.5	52.5	53.33
	r	4.5	5	6	5.17

		Evaluation Lot 1	Evaluation Lot 2	Evaluation Lot 3	Average Value
fr	Average	450.8	449.72	449.86	450.13
	$\sigma_{n-1}$	0.90	1.03	1.10	1.01
	max.	454.00	454.00	454.50	454.17
	min.	448.00	447.00	447.00	447.33
	r	6.00	7.00	7.50	6.83

**Table 2**  
**Comparative Example**

		Evaluation Lot 1	Evaluation Lot 2	Evaluation Lot 3	Evaluation Lot 4	Evaluation Lot 5	Average Value
df	Average	56.44	56.22	56.41	56.69	56.31	56.41
	$\sigma_{n-1}$	2.19	1.97	2.09	2.01	1.97	12.04
	max.	63.00	62.50	62.50	63.00	63.00	62.80
	min.	52.50	51.50	52.50	53.00	53.00	52.80
	r	10.50	11.00	10.00	10.00	10.00	10.30

		Evaluation Lot 1	Evaluation Lot 2	Evaluation Lot 3	Evaluation Lot 4	Evaluation Lot 5	Average Value
fr	Average	448.99	448.69	448.63	448.96	447.88	448.63
	$\sigma_{n-1}$	1.37	1.41	1.19	1.29	1.15	1.28
	max.	452.50	452.00	451.50	452.00	450.50	451.70
	min.	444.50	443.50	444.50	445.00	444.50	444.40
	r	8.00	8.50	7.00	7.00	6.00	7.30

Tables 1 and 2 show that the average value of the standard deviation  $\sigma_{n-1}$  of df, is 2.04kHz in the first example and 0.91kHz in the Comparative Example. The first example has approximately half of the variation in a polarization degree df of the Comparative Example. Moreover, the average value of the standard deviation  $\sigma_{n-1}$  of a resonance frequency fr is 1.28kHz in the first example and 1.01kHz in the Comparative Example. Thus, the variation in the resonance frequency fr has been reduced by about 30%.

#### Second Example

Fig. 12 shows a manufacturing process of a length mode piezoelectric resonator according to a second example of preferred embodiments of the present invention.

In this example, a plurality of operations of the first polarization are performed in the state of a block 1 (referring to (a) and (b) of Fig. 12) and the direction of polarization was reversed through the entire block 1.

In this case, the difference  $\Delta P2$  between a center section and an edge portion decreases as the polarization is reversed as shown in Fig. 4. Therefore, when the block is cut into a rectangle 1A in (c), the polarization degree variation (concave distribution) between rectangles 1A and within a rectangle 1A decreases. Therefore, when a secondary polarization is performed in (c), a concave polarization degree distribution of the piezoelectric layer 1c is equalized.

In addition, the process of (b) is not restricted to a single process, but may be performed a plurality of times.

Table 3 shows the lot variation in the polarization degree  $df$  and the resonance frequency  $fr$  of the second example.

As shown in Table 3, the average value of the standard deviation  $\sigma_{n-1}$  of a polarization degree  $df$  is 0.85kHz and the average value of the standard deviation  $\sigma_{n-1}$  of a resonance frequency  $fr$  is 0.96kHz. Thus, the second example further reduces the variation.

**Table 3**  
**Second Example**

		Evaluation Lot 1	Evaluation Lot 2	Evaluation Lot 3	Average Value
$df$	Average	53.93	53.44	54.03	53.80
	$\sigma_{n-1}$	0.96	0.71	0.87	0.85
	max.	58.00	55.00	57.50	56.83
	min.	50.00	51.50	52.00	51.17
	$r$	8.00	3.50	5.50	5.67

		Evaluation Lot 1	Evaluation Lot 2	Evaluation Lot 3	Average Value
$fr$	Average	449.15	448.93	449.06	449.05
	$\sigma_{n-1}$	0.85	0.97	1.06	0.96
	max.	451.50	452.00	451.50	454.17
	min.	446.00	446.00	446.50	446.17
	$r$	5.50	6.00	5.00	5.50

### Third Example

Fig. 13 shows a manufacturing process of a length mode piezoelectric resonator according to a third example of preferred embodiments of the present invention.

The electro-conductive paste for internal electrodes that includes silver, palladium, an organic binder, or other suitable material, is applied on one side of a green sheet including a piezoelectric ceramic. These are alternately laminated and are integrally baked at about 1200 degree C, and then the multi-layered piezoelectric body 1 having approximate dimensions of 20 mm x 30 mm x 3.9 mm in a block form was produced. In addition, the side surface electrodes 3 and 4 for leading out an internal electrode are alternately provided on a side surface of the multi-layered piezoelectric body 1. A DC electric field was applied between the side surface electrodes 3 and 4 and a first polarization



was performed (referring to (a) of Fig. 13).

The conditions of the first polarization are as follows: electric-field is 1.5 kv/mm, polarization time is 10 min., and retention-temperature is about 70 degree C. After that, an aging process was performed at about 150 degree C for 1 hour.

Next, the block-like multi-layered piezoelectric body 1 was cut in a vertical direction using a dicer to produce a substantially rectangular shape. The DC electric field is applied to the cut rectangle 1A via the side surface electrodes 3 and 4, and a secondary polarization was performed (referring to (b) of Fig. 13). At this time, the direction of the voltage application to the rectangle 1A was in the reversed direction to that of the first polarization. The degree of polarization of each rectangle 1A was uniformly set to a desired value by the control of the polarization time.

The conditions of the secondary polarization are set as follows: electric-field is 1.5 kv/mm, and polarization temperature is about 70 degree C. A polarization time is controlled to achieve the desired polarization degree (polarization degree of the rectangle responded in polarization degree  $df=55\text{kHz}$  of a length mode element). After that, the aging process was performed at about 250 degree C for about 1 hour.

The range of the secondary polarization is determined as follows: A range in which a remanent polarization degree  $Pr_2$  wherein the difference  $df$  between the resonance frequency and the antiresonance frequency of the length oscillation mode of the rectangle after a secondary polarization is converted into electro-mechanical-coupling-coefficient  $K$  does not exceed a remanent polarization degree  $Pr_1$  wherein the difference  $DF$  between the resonance frequency and the antiresonant frequency of the length oscillation mode of the block after the first polarization is converted into electro-mechanical-coupling-coefficient  $K$ , that is,  $Pr_1 \geq Pr_2$ .

Every other electrode exposed at a side surface of the rectangle 1A is coated with an insulating material after the secondary polarization, and a silver electrode was provided thereon. The dicer cuts this, and a 1.5 mm x 1.5 mm x 3.8 mm length mode piezoelectric resonator 1B was obtained. The structure of this piezoelectric resonator 1B is the same as that of the piezoelectric resonator of the first example.

The frequency characteristic of the impedance in piezoelectric resonator of the third example and in the piezoelectric resonator of Comparative Example

is measured.  $df=55\text{kHz}$  value was obtained as a difference between a resonance frequency and an antiresonance frequency.

Table 4 shows lot variations of the polarization degree  $df$  and the resonance frequency  $fr$  in the third example.

As shown in Table 4, the average value of the standard deviation  $\sigma_{n-1}$  of a polarization degree  $df$  is  $1.03\text{kHz}$  and the average value of the standard deviation  $\sigma_{n-1}$  of a resonance frequency  $fr$  is  $0.92\text{kHz}$ . The variation in a polarization degree  $df$  is substantially greater than in the first example. However, the variation in a resonance frequency  $fr$  is the smallest of the three examples.

**Table 4**  
**Third Example**

		Evaluation Lot 1	Evaluation Lot 2	Evaluation Lot 3	Average Value
$df$	Average	54.72	54.91	55.91	55.18
	$\sigma_{n-1}$	0.96	0.99	1.14	1.03
	max.	56.53	55.82	58.15	56.83
	min.	51.61	50.44	52.50	51.52
	$r$	4.91	5.38	5.65	5.31

		Evaluation Lot 1	Evaluation Lot 2	Evaluation Lot 3	Average Value
$fr$	Average	450.62	449.56	452.26	450.81
	$\sigma_{n-1}$	0.71	0.93	1.12	0.92
	max.	451.90	450.98	454.00	452.29
	min.	447.93	446.20	448.68	447.60
	$r$	3.97	4.78	5.32	4.69

The polarization method of this invention is not limited to the above-described examples of preferred embodiments.

For instance, the secondary polarization was performed only once to the piezoelectric body 1A of a rectangular shape in Figs. 11 to 13. However, the secondary polarization may be repeated several times. In other words, the direction of an electric field may be reversed and reversal of a direction of polarization may be repeated several times.

Moreover, when the mass-production property and a polarization degree distribution are considered, a first polarization is performed to a block-like multi-layered piezoelectric body and a secondary polarization is performed to the multi-layered piezoelectric body having a substantially rectangular shape.

However, a first polarization and a secondary polarization may be performed on a block-like multi-layered piezoelectric body, and a first polarization and a secondary polarization may be performed to the multi-layered piezoelectric body having a substantially rectangular shape.

In addition, in the first and second examples (Figs. 5, 6, 11 and 12), after a secondary polarization, the remaining polarization degree of the piezoelectric layer having a direction of polarization that is reversed was temporarily set to  $Pr_2$  and was explained. This is intended to simplify the understanding of these examples. In actuality, the entire (the piezoelectric layer in which the repolarization layer and the direction of polarization are reversed is included) rectangle after a secondary polarization is excited by the length oscillation mode and the remanent polarization degree thereof is set to  $Pr_2$ .

As described above, according to a preferred embodiment of the present invention, after performing the first polarization which uniformly polarizes the multi-layered piezoelectric body in the thickness direction, when the secondary polarization which reverses the direction of polarization of only the piezoelectric layer of one side of an internal electrode is performed, because the secondary polarization is performed in a range in which the remanent polarization degree  $Pr_2$  after the secondary polarization in the piezoelectric layer having a direction of polarization that is reversed does not exceed the remaining polarization degree  $Pr_1$  after the first polarization, the polarization degree distribution of the piezoelectric layer having a direction of polarization that is reversed is convex-shaped or flat. Therefore, since the polarization degree distribution of the piezoelectric layer having a direction of polarization that is reversed is convex-shaped or flat, and a polarization degree distribution of the piezoelectric layer having a direction of polarization that is not reversed is concave, an approximately uniform polarization degree distribution is obtained for the overall multi-layered piezoelectric body. As a result, when cutting and using a multi-layered piezoelectric body, that usable portion greatly increases, and thus the yield greatly improves.

According to another preferred embodiment of the present invention, after performing the first polarization which polarizes the piezoelectric layers on both sides of the internal electrode in opposite directions, when the secondary polarization which reverses the direction of polarization of the piezoelectric layers on both sides of the internal electrode is performed, because a secondary polarization is performed in a range in which the remanent

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